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Dynamics of FLCs with a tilted bookshelf structure using time-resolved FTIR spectroscopy

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Time-resolved FTIR spectroscopy with microsecond resolution was applied to the switching process of FLCs using an IR polarizer attached to a microscope whose axis was set along the aligning direction (direction of SiO flux). A tilted bookshelf geometry with a layer tilt angle of approximately 34° was obtained by alignment using obliquely evaporated SiO layers in a CaF_2 cell with about $1.7 \mu\text{m}$ gap. Six kinds of FLC mixtures were prepared by mixing 5 wt % of a chiral molecule with host mixtures of phenylpyrimidines. By monitoring an absorption band at 1432 cm^{-1} , assigned to vibrational modes of the pyrimidine skeleton, it was found that the molecular long axis either rotates along the upper side or along the lower side of the cone during switching. Moreover, we investigated the relationship between the rotational direction of the directors and the stability of the dark level transmission during multiplexing. The dark level transmission during rotation along the upper side of the cone may fluctuate less than that during rotation along the lower side of the cone. We found that the bias stability of the dark level transmission is closely related to the direction of the rotation along the cone. The direction of the rotation of FLCs along the cone was therefore found to play an important role in realizing high contrast ratios during multiplexing.

1. Introduction

Displays based on ferroelectric liquid crystals (FLCs) using the chiral smectic C (S_C^*) phase are known as surface-stabilized ferroelectric liquid crystal displays (SSFLCDs) [1]. FLCs have been studied with the aim of realizing the next generation of high resolution flat panel displays, due to their excellent electro-optical properties such as fast response, the memory effect and wide viewing angle. The response times and contrast ratios, however, are not yet sufficiently high for video rate displays.

In order to realize fast switching speeds and high contrast ratios in FLC displays, it is important to clarify the dynamics of the FLCs during switching. We reported previously that a novel molecular parameter α is in good correlation with response times and cone angles. Also, the parameter α is closely related to the length of rigid and/or flexible parts of chiral molecules [2].

In particular, contrast ratios during multiplexing are important for realizing a display with passive matrix driving. The contrast ratios during multiplexing are generally considered to relate to dielectric anisotropy of FLC mixtures, that is, in the FLC mixtures having negative dielectric anisotropy, the position of the director is stabilized by the dielectric torque, proportional to $\Delta\epsilon E^2$ so that the dark level and the bright level transmission are

maintained [3]. However, FLC molecules and their mixtures designed to have negative dielectric anisotropy have not always exhibited high dark level stability.

In this study, we investigated the rotation behaviour of FLC molecules on the cone surface during switching utilizing a time-resolved FTIR spectroscopic method, and found that the rotational directions on the cone govern the stability of dark levels on exposure to data pulses.

2. Experimental

2.1. FLC mixtures

Chiral molecules were employed as shown in figure 1. In order to use FLCs with large spontaneous polarizations, fluorine was introduced into the chiral carbon in the chiral end group. The core groups are biphenyl (C8 LPS, C10 LPS, C10F) and aromatic esters (FPB, FFBB, FNB). In order to use FLCs with negative dielectric anisotropy, two of the hydrogen atoms of the biphenyl core were substituted with fluorines [4].

FLC mixtures were prepared by mixing the above chiral molecules with host mixtures of phenylpyrimidines, as shown in figure 2. For example, the C10F mixture contains 5 wt % of C10F and the remainder consists of equal amount of the two host molecules. The phase sequence of this mixture is $\text{Cr } 14 \text{ S}_\text{C}^* 53 \text{ S}_\text{A} 59 \text{ N}^* 66 \text{ I}$. The phase sequences of the others are similar to that of the C10F

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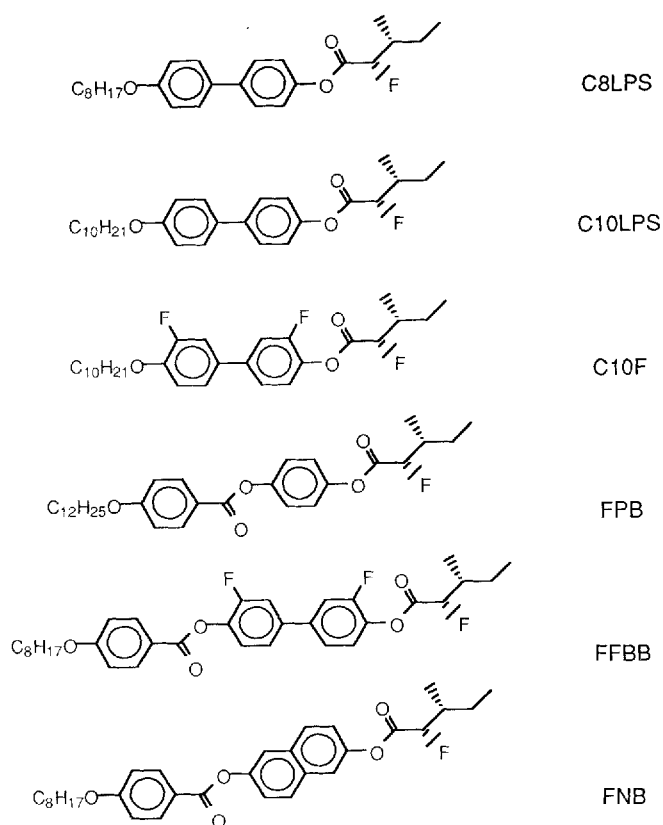


Figure 1. Structure of the chiral molecules.

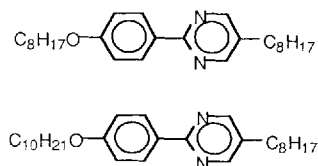


Figure 2. Structure of the host molecules.

mixture. The measurements described in this paper were therefore all carried out at 40°C: equal to $T_{AC} - 13^\circ\text{C}$.

2.2. FLC cell fabrication

Homogeneously aligned cells were fabricated using an obliquely evaporated SiO alignment layer (50 nm) on CaF₂ substrates with transparent sputtered ITO electrodes (50 nm) and with a gap of approximately 1.7 μm in the anti-parallel configuration [2, 5]. The cells were filled with the FLC mixtures at a temperature above the clearing point *in vacuo*, and then cooled gradually down ($-2^\circ\text{C min}^{-1}$) to room temperature.

2.3. Electro-optic measurements

The electro-optic measurements were made by using a polarized optical microscope, a wave function generator,

a digital oscilloscope, a photo multiplier (Hamamatsu Photonics R-928) and a filtered halogen light source (with a centre wavelength of 530 nm and a half width of 60 nm). In our electro-optic measurements, the polarizer axis was set perpendicular to the analyser axis, and parallel or perpendicular to an optical axis in the memory states. The optical cone angles on applying an electric field, $2\theta_E$, and the cone angles in the memory state, $2\theta_M$, are defined as the difference in the extinction angle between the switching states.

The signal pattern used for evaluating the response time was a 200 ~ 500 Hz square wave, 30 V_{pp} at 40°C. The rise time (τ_r) is defined as the time required for the optical transmission to change from 10 to 90 per cent and the delay time (τ_d) is defined as the time required for the optical transmission to change from 0 to 10 per cent, where the maximum optical transmission for each sample is normalized as 100 per cent.

The texture change of the FLCs in a 100 μm square area was monitored versus time during switching using an image intensified CCD video camera system (Hamamatsu Photonics C-4053H) with microsecond resolution attached to a polarized microscope.

2.4. FTIR measurements

Asynchronous time-resolved FTIR spectroscopy was performed using a Jeol JIR-6500 system with a time-resolution attachment and a microscope unit imaging 100 μm square [6]. The gate width was 10 μs and the interval was either 20 or 40 μs . The IR polarizer was set along the aligning direction (direction of SiO flux). FLCs were switched using a 600 Hz square wave, 30 V_{pp} at 40°C.

3. Results and discussion

3.1. Microscopic observation

In the FLC cells with an anti-parallel configuration, zigzag defect free, monodomain textures were observed under crossed polarizers. This corresponds to the twisted state, showing a bluish grey colour [7]. On applying an electric field, those textures were, however, changed from the twisted state to a uniform one, and the cells exhibited good bistability, except of for the FFBB and FNB systems, without the application of an applied voltage. Contrast ratios above 40:1 were achieved in the visible region.

The texture in the observed region was uniformly changed during switching without a growth of the reversed domain along the cell surface. It is considered that the movement of the FLC molecules with time during switching can be evaluated using a typical cone model for the FLC switching.

3.2. Geometry of the layer structure of the S_C^* phase in the cells

The layer spacings in the smectic C^* phase obtained from Bragg's angle $2\theta_B$ were almost proportional to the length of chiral molecules in the FLC mixtures that have been reported [2]. Those FLC cells have a tilted bookshelf geometry for the S_C^* layers with a layer tilt angle δ in the range of about 26° to 36° measured by using X-ray diffraction. A systematic dependence of the layer tilt angles among the chiral molecules studied was not observed.

3.3. Rotation behaviour of the FLC molecules

Dichroic ratios of 8 or above were observed for the infrared absorption bands of phenylpyrimidines core. The liquid crystal molecules were well aligned in those cells. Time-resolved FTIR spectra of the C10F and FPB systems are shown in figures 3 and 4. The absorption bands observed in the region from 1150 to 1650 cm^{-1} are mainly attributable to the bands from phenylpyrimidines core group. These bands show similar absorbance behaviour

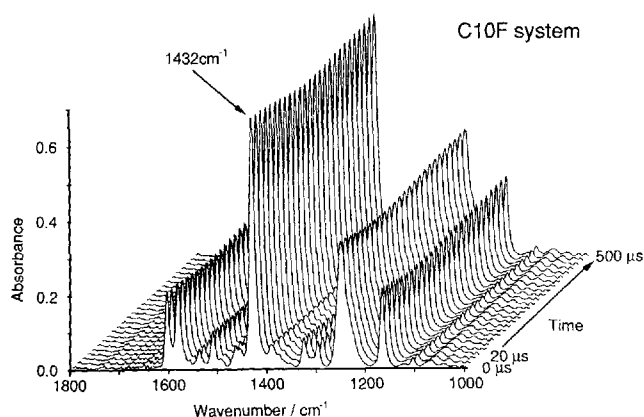


Figure 3. Time-resolved FTIR spectra of the C10F system. $10\text{ }\mu\text{s}$ gate width and $20\text{ }\mu\text{s}$ step increments.

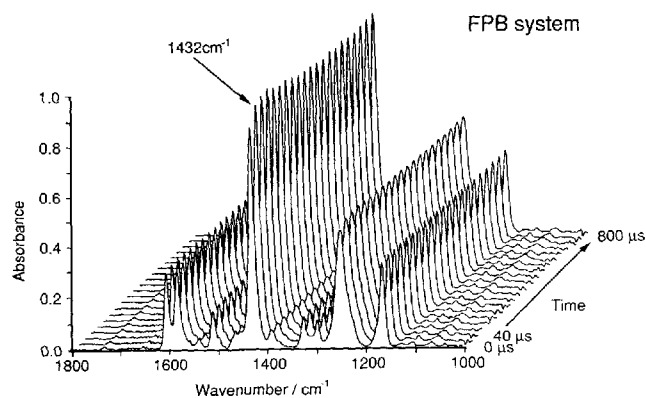


Figure 4. Time-resolved FTIR spectra of the FPB system. $10\text{ }\mu\text{s}$ gate width and $40\text{ }\mu\text{s}$ step increments.

with time. In particular, the absorption band at 1432 cm^{-1} is assigned to vibrational modes of the pyrimidine skeleton, parallel to the molecular long axis. Peak absorbance changes with time at 1432 cm^{-1} for the C10F and FPB systems are shown in figure 5. These absorbances were normalized to the maximum values.

In the C10F system, the absorption band at 1432 cm^{-1} decreases until $t = 200\text{ }\mu\text{s}$ and then increases thereafter. Here $t = 0$ corresponds to the time of the polarity switch of the bipolar switching pulse. However, in the FPB system, the absorption band at 1432 cm^{-1} increases until $t = 120\text{ }\mu\text{s}$ and then decreases thereafter. In particular, observing the changes in the absorption intensity of tilted smectic layers makes it easy to elucidate the motion of liquid crystal director. If we assume a cone model of FLC

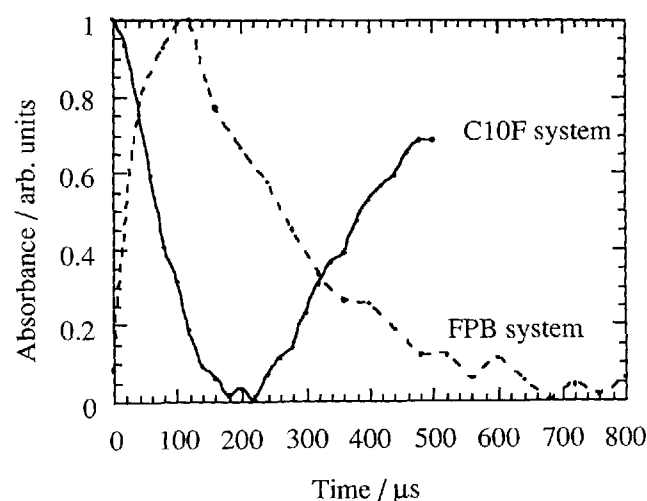


Figure 5. Peak absorbance changes with time at 1432 cm^{-1} for the C10F and FPB systems. These absorbances were normalized to the maximum value.

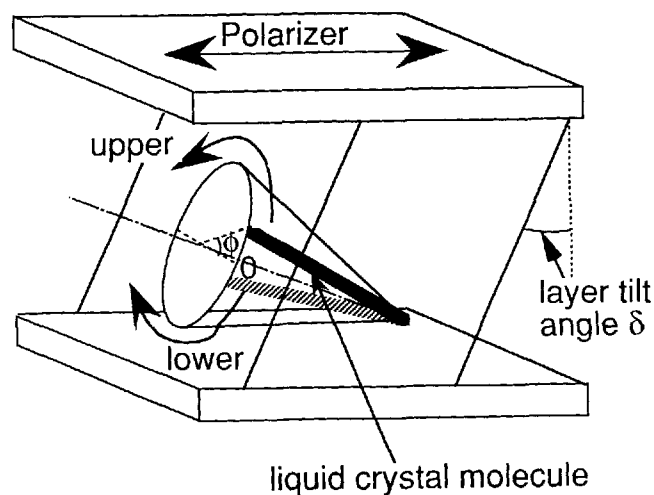


Figure 6. Model of the director rotation in the tilted bookshelf geometry during the time-resolved FTIR spectroscopy.

switching in a tilted bookshelf geometry, as shown in figure 6, we can determine the director rotation as follows. The intensity change in the absorbance at 1432 cm^{-1} for the C10F system is concave, thereby indicating that the molecular long axis rotates along the upper side of the cone. On the other hand, the absorbance profile for the FPB system is convex, indicating a rotation along the lower side of the cone. The other systems exhibit rotation along the upper side of the cone. It is interesting that only 5 wt % of chiral molecules dominate the switching of the FLC.

If we assume a cone model for the FLC switching in the tilted bookshelf geometry as illustrated in figure 7, the optical transmission as a function of an azimuthal angle

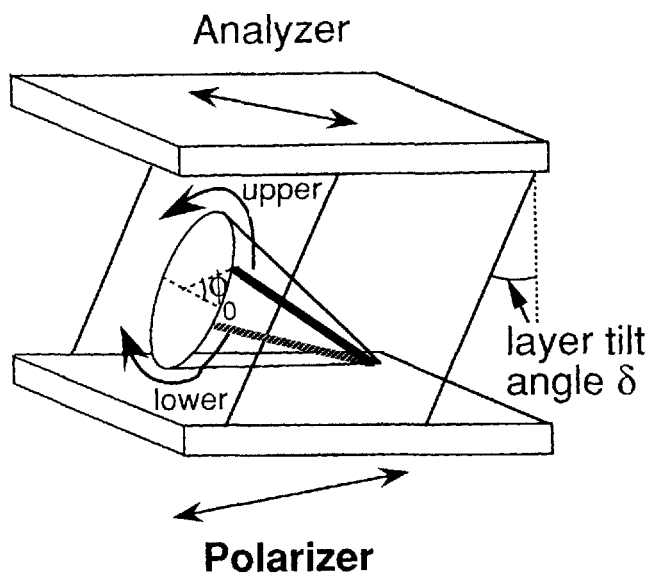


Figure 7. Geometry used for the calculation of the optical transmission change.

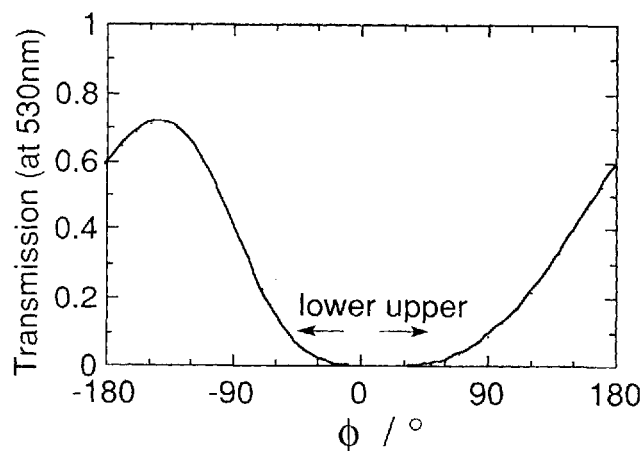


Figure 8. The calculated optical transmission change at 530 nm with an azimuthal angle of the director, ϕ in the C10F system.

ϕ may be calculated by considering the layer tilt, the cone angle and the birefringence using Berreman's 4×4 -matrix formulation [8]. The polarizer axis is set along the director at $\phi = 0$ and is perpendicular to the analyser axis.

The optical transmission at 530 nm as a function of an azimuthal angle ϕ in the C10F system is shown in figure 8. The birefringence, 0.13, used for calculation was obtained from the phenylpyrimidine mixture shown in figure 2.

A positive azimuthal angle, $\phi > 0$, shows a rotation along the upper side of the cone. On the other hand, a negative azimuthal angle, $\phi < 0$, shows a rotation along the lower side of the cone. When the dark level transmission is below 0.01, the contrast ratio becomes larger than 60:1. The transmission is less than 0.01 until an azimuthal angle of 50° when rotated along the upper side of the cone and until -20° when rotated along the lower side of the cone. It is estimated that the dark level transmission when rotated along the upper side of the cone may fluctuate less than in the reverse rotation around an azimuthal angle of 0° . That is to say, the dark level transmission during multiplexing should be governed by the direction of the rotation along the cone.

3.4. Stability of the dark level during multiplexing

The optical transmission as a function of time on the application of multiplexing square wave pulses ($180\ \mu\text{s}$) in the C10F system are shown in figures 9(a)–(c) showing the stability of the dark level transmission.

The signal used for evaluating the stability of the dark level transmission when the device is multiplexed consists of a series of bipolar pulses that consist of the scan pulses and bias pulses that arise when the data pulses are applied. This is to avoid electrolytic and charge accumulation (displacement) effects which would result from a d.c. offset. The amplitudes of the write and reset pulses were 15 V, while those of the data pulses were from 0 to 7.5 V. For example, the bias ratio during the period of blanking becomes 2:1 when the data pulse is 7.5 V. The stability of the dark level transmission when the cell was multiplexed was evaluated as the ratio of the maximum transmission of the dark level with the application of data pulses divided by the bright level in the memory states without data pulses.

In the C10F system, switching between the memory states can easily be realized by a short square pulse ($180\ \mu\text{s}$, 15 V), which is the minimum pulse width for latching. Bright level transmission is varied by the application of data pulses during non-selecting, but dark level transmission is maintained with data pulses of up to 4.5 V. The contrast ratio is defined as the ratio between the average bright and dark level transmission during the period of non-selecting. The contrast ratio as a function of the amplitude of the data pulses in the C10F system is

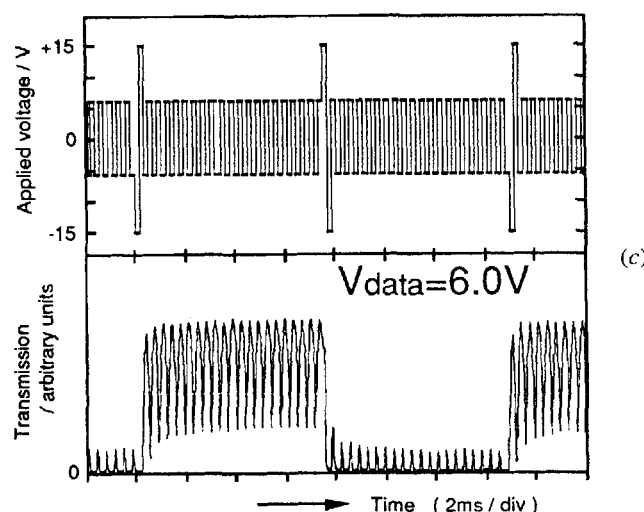
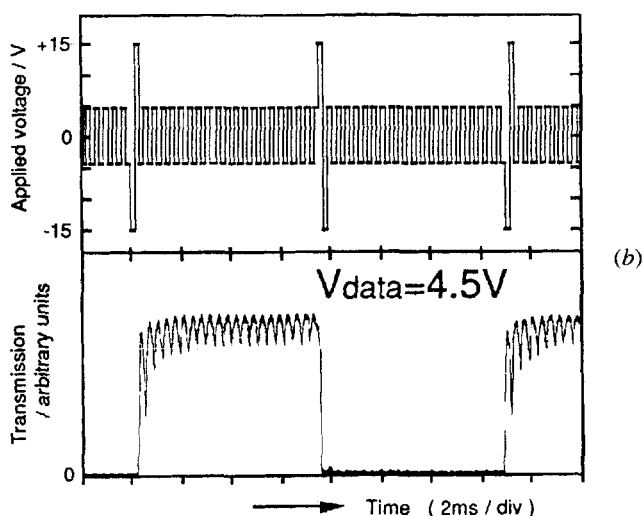
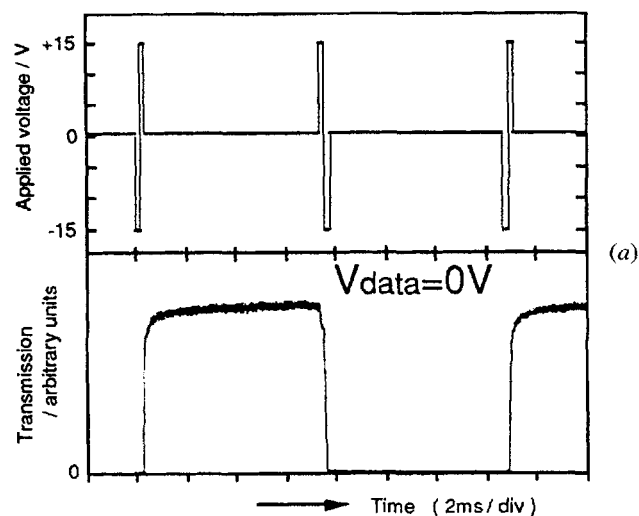


Figure 9. The optical transmission as a function of time during the application of multiplexing square wave pulses (180 μ s) in the C10F system. The amplitude of the data pulses are 0 V (a), 4.5 V (b) and 6.0 V (c), respectively.

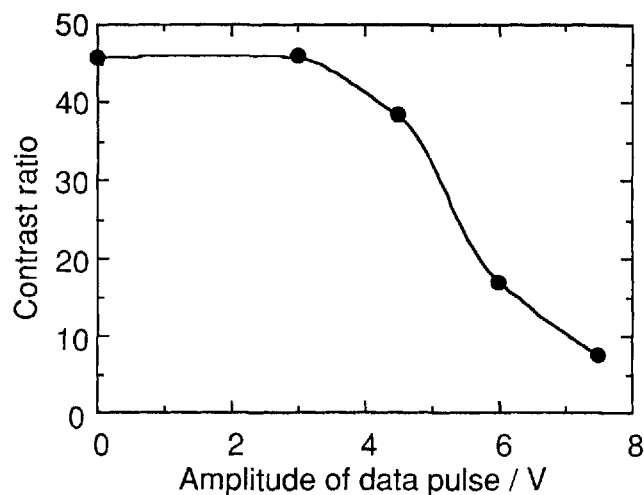


Figure 10. The contrast ratio as a function of the amplitude of the data pulses in the C10F system.

shown in figure 10. It is maintained until 4.5 V and decreases thereafter. The degradation in the contrast ratio is not only due to the fluctuation of the bright level, but also to that of dark level. In particular, the contrast ratio is greatly influenced by the transmission of the dark level. We therefore noted the role of dark level transmission during multiplexing.

3.5. Relationship between the rotational direction of the directors and the stability of the dark level

The relationship between the ratios of cone angle θ_M/θ_E and the ratios of response time $\tau_{0-10\%}/\tau_{0-90\%}$ is shown in figure 11. The ratio $\tau_{0-10\%}/\tau_{0-90\%}$ represents the stability of the dark level at the full cone angle, while the fluctuation of the dark level during multiplexing represents the stability of the memory state. The ratio $\tau_{0-10\%}/\tau_{0-90\%}$ is proportional to θ_M/θ_E . It is therefore estimated that the stability of the dark level at the full cone angle is degraded by the effect of the twisted state. In the C10F, C10LPS and FPB systems, the cells show good bistability in the memory state, because the value of θ_M/θ_E equals one. However, the value of $\tau_{0-10\%}/\tau_{0-90\%}$ for the FPB system is too small to stabilize the dark level at the full cone angle.

The stability of the dark level transmission during multiplexing as a function of amplitude of the data pulses in various systems is shown in figure 12. For data pulses of less than 3 V, the dark levels in the biphenyl core systems (C8 LPS, C10 LPS, C10F) tend to be more stable than in the aromatic ester cores systems (FPB, FFBB, FNB) below 3 V of data pulses. In the C10F molecule two of the hydrogen atoms of the biphenyl core of the C10 LPS molecule are substituted by fluorines. It is considered that the C10F system is more stable than the C10 LPS system because of the negative dielectric anisotropy of the former. In aromatic ester core systems, the stability of the dark

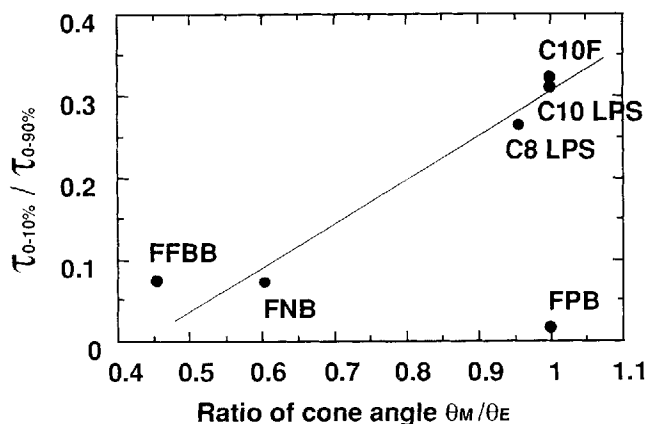


Figure 11. The relationship between the ratios of the cone angle θ_M/θ_E and the ratios of response time $\tau_{0-10\%}/\tau_{0-90\%}$ in various systems. the FFBB and FNB systems are still in twisted states after the application of an electric field.

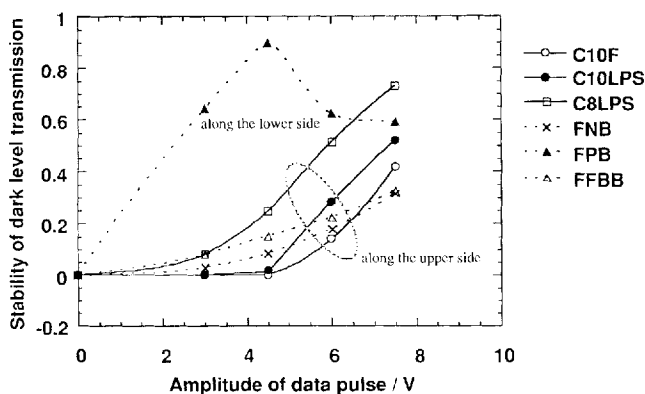


Figure 12. The stability of the dark level transmission during multiplexing as a function of the amplitude of the data pulses in various systems. The director of the FPB system only rotates along the upper side of the cone.

level of the FPB system is the most unstable among the others, so that a passive matrix display can't be realized by using the FPB mixture under above conditions.

Only the FPB system among the FLCs which showed good bistability in the memory state showed a rotation along the lower side of the cone. The large fluctuation in the dark level transmission in the FPB system is explained as above.

Consequently, it is shown that the stability of the dark level during multiplexing is governed by a rotational direction of the directors along the cone. We believe that the rotational direction of the directors may be influenced by the initial torque on the chiral molecules upon the application of an electric field. The initial torque may be determined by the spontaneous polarization P_s , dielectric

anisotropy $\Delta\epsilon$, and an azimuthal angle ϕ in the memory state, which may be dominated by the cone geometry such as the cone angle, layer tilt angle and pretilt angle.

4. Summary

Time-resolved FTIR spectroscopy with microsecond resolution was used to investigate six kinds of FLC systems with good alignment and an ideal tilted bookshelf geometry. It was clarified that the molecular long axis rotates along the upper side of the cone in the C8 LPS, C10 LPS, C10F, FFBB, and FNB systems, while along the lower side of the cone in the FPB systems. It is interesting that the addition of only 5 wt% of chiral molecules dominates the switching properties of these FLC molecules. We believe that the rotational direction of the directors may be decided by the initial torque on the chiral molecules upon application of an electric field.

The contrast ratios during multiplexing have been considered to be related to the dielectric anisotropy of the FLC mixtures. However, we found that a relationship exists between the rotational direction of the directors and the stability of the dark level transmission during multiplexing, so that the dark level fluctuation is governed by the direction of the rotation along the cone. The fluctuation of the dark level transmission during rotation along the upper side of the cone may be smaller than that of the transmission when the rotation is along the lower side of the cone. Therefore, the direction of the rotation of the FLCs along the cone was found to be the crucial factor in realizing high contrast ratios during multiplexing.

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